

What is claimed is:

1. A non-aqueous electrolyte secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolyte, wherein the negative electrode contains graphite in which  $G_s$  denoted by the following formula in the surface enhanced Raman spectrum is 10 and below.

$$G_s = H_{sg}/H_{sd}$$

(where,  $H_{sg}$  is the height of a signal having a peak within the range of 1580  $\text{cm}^{-1}$  to 1620  $\text{cm}^{-1}$ , both inclusive, and  $H_{sd}$  is the height of a signal having a peak within the range of 1350  $\text{cm}^{-1}$  to 1400  $\text{cm}^{-1}$ , both inclusive)

112? 2. A non-aqueous electrolyte secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolyte, wherein the negative electrode contains graphite having at least two peaks on a defferential thermogravimetric curve obtained by thermogravimetric analysis in an airflow.

DTA  
Differential Thermal Analysis

3. A non-aqueous electrolyte secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolyte, wherein the negative electrode contain graphite with a saturated tapping density of 1.0 g/ $\text{cm}^3$  and more.

4. A non-aqueous electrolyte secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolyte, wherein the

negative electrode contains graphite with a packing characteristic index of 0.42 and more.

5. A non-aqueous electrolyte secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolyte, wherein the negative electrode formed by pressing and contains graphite with a specific surface area after pressing being 2.5 times and below of that before pressing.

6. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein interlayer spacing distance [d (002)] crystal planes parameter of the graphite in the negative electrode is 0.3363 nm and below.

7. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the starting temperature of weight reduction by oxidation of the graphite in the negative electrode obtained by thermogravimetric analysis in an airflow is 300 °C and more.

8. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the graphite in the negative electrode has at least two peaks on the differential thermogravimetric curve obtained by thermogravimetric analysis in an airflow, and the reform rate obtained from the differential thermogravimetric curve lies within the range of 1 to 38, both inclusive.

9. A non-aqueous electrolyte secondary battery as claimed in claim 1,

wherein the saturated tapping density of the graphite in the negative electrode is  $1.0 \text{ g/cm}^3$  and more.

10. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the packing characteristic index of the graphite in the negative electrode is 0.42 and more.

11. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the negative electrode is formed by pressing and a specific surface area of the graphite in the negative electrode after pressing is 2.5 times and below compared to that before pressing.

12. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the graphite in the negative electrode has a rhombohedral structure.

13. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein the positive electrode is formed containing a lithium composite oxide expressed by  $\text{LiM}_x\text{O}_y$  (where M denotes at least one element selected from the group consisting of Co, Ni, Mn, Fe, Cr, Al and Ti).

14. A non-aqueous electrolyte secondary battery as claimed in claim 1, wherein each of the negative electrode and the positive electrode has a structure in which a mixture containing an active material is formed on both

sides of a band-shaped collector, and the battery is formed by stacking the negative electrode and the positive electrode with a separator made of a microporous film interposed therebetween, being spirally rolled a number of times.

15. A method of preparing a carbon-based material for a negative electrode including steps of:

mixing a coating material made of one of pitch containing free carbon, pitch with a quinoline insoluble matter content of 2 % and more, or polymer with a carbon-based material made of at least either one of mesocarbon microbeads grown at a temperature within the range of the formation temperature to 2000 °C, both inclusive, and a carbon material; and

graphitizing the carbon-based material with which the coating material is mixed.

16. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15 including a step of: applying a heat treatment to the carbon-based material in an oxidizing atmosphere before graphitization.

17. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15 including a step of: performing an oxidation treatment on the carbon-based material by one or more methods of an acid treatment, an ozone treatment or air oxidation before mixing the carbon-

based material with the coating material.

18. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein pitch comprising carbon black is used as the coating material.

19. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein natural graphite having a rhombohedral structure is used as the carbon-based material.

20. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein graphite powder with the tapping density of  $1.0 \text{ g/cm}^3$  and more in tapping 40 times is used as the carbon-based material.

21. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein natural graphite with the tapping density of  $0.9 \text{ g/cm}^3$  in tapping 20 times is used as the carbon-based material and the graphitization is performed at a temperature within the range of  $200^\circ\text{C}$  to  $2300^\circ\text{C}$ , both inclusive.

22. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein natural graphite with the tapping density of  $0.9 \text{ g/cm}^3$  in tapping 40 times, on which pressing is performed, is

used as the carbon-based material.

23. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein each of a negative electrode and a positive electrode has a structure in which a mixture containing an active material is formed on both sides of a band-shaped collector, and the battery is formed by stacking the negative electrode and the positive electrode with a separator made of a microporous film interposed therebetween, being spirally rolled a number of times.

24. A method of preparing a carbon-based material for a negative electrode as claimed in claim 23, wherein the positive electrode is formed to contain a lithium composite oxide expressed by  $\text{LiM}_x\text{O}_y$  (where M denotes at least one element selected from the group consisting of Co, Ni, Mn, Fe, Cr, Al and Ti).

25. A method of preparing a carbon-based material for a negative electrode as claimed in claim 22, wherein the pressing is performed with a pressure of 1 MPa and more.

26. A method of preparing a carbon-based material for a negative electrode including steps of:

applying a heat treatment in an oxidizing atmosphere on a carbon-based material made of at least either one of mesocarbon microbeads grown

at a temperature within the range of the formation temperature to 2000 °C, both inclusive, and a carbon material; and

graphitizing the carbon-based material.

27. A method of preparing a carbon-based material for a negative electrode as claimed in claim 26, wherein natural graphite having a rhombohedral structure is used as the carbon-based material.

28. A method of preparing a carbon-based material for a negative electrode as claimed in claim 26, wherein graphite powder with the tapping density of 1.0 g / cm<sup>3</sup> and more in tapping 40 times is used as the carbon-based material.

29. A method of preparing a carbon-based material for a negative electrode as claimed in claim 26, wherein each of a negative electrode and a positive electrode has a structure in which a mixture containing an active material is formed on both sides of a band-shaped collector, and the battery is formed by stacking the negative electrode and the positive electrode with a separator made of a microporous film interposed therebetween, being spirally rolled a number of times.

30. A method of preparing a carbon-based material for a negative electrode as claimed in claim 29, wherein the positive electrode is formed to contain a lithium composite oxide expressed by  $\text{LiM}_x\text{O}_y$  (where M denotes at

least one element selected from the group consisting of Co, Ni, Mn, Fe, Cr, Al and Ti).

31. A method of preparing a carbon-based material for a negative electrode including steps of:

mixing a coating material made of one of pitch containing free carbon, pitch with a quinoline insoluble matter content of 2 % and more, or polymer with a carbon-based material made of at least either one of mesocarbon microbeads grown at a temperature within the range of the formation temperature to 2000 °C, both inclusive, and a carbon material; and

graphitizing the carbon-based material to which the coating material is mixed;

wherein the step of mixing the coating material with the carbon-based material includes a step of applying a heat treatment to graphite particles in an inert atmosphere where more than a specific concentration of an organic substance is diffused.

32. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein the method is used in a manufacturing process of a secondary battery where each of a negative electrode and a positive electrode has a structure in which a mixture containing an active material is formed on both sides of band-shaped collector and the battery is formed by stacking the negative electrode and the positive electrode with a separator made of a microporous film interposed



therebetween, being spirally rolled a number of times.

33. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein a benzenoid compound having a structure including at least one or more benzene rings is used as the organic substance.

34. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein a benzenoid compound having a structure including a bond of oxygen is used as the organic substance.

35. A method of preparing a carbon-based material for a negative electrode as claimed in claim 33, wherein either a nonbenzenoid compound, or a mixture of a nonbenzenoid compound and a benzenoid compound is contained as the organic substance.

36. A method of preparing a carbon-based material for a negative electrode as claimed in claim 34, wherein a compound in which a carbon atom (C) is replaced with another element is used as the benzenoid compound.

37. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein a benzenoid compound having at least one or more structures in which a carbon atom (C) is replaced with at

least one element selected from the group consisting of sulfur (S), nitrogen (N) and phosphorus (P) is used as the benzenoid compound.

38. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31 including a step of forming the graphite particles by grinding artificial graphite or natural graphite.

39. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein an oxidation treatment is performed on the graphite particles by one or more methods of an acid treatment, an ozone treatment or air oxidation before the heat treatment.

40. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein natural graphite having a rhombohedral structure is used as the graphite particles.

41. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein graphite powder with the tapping density of  $1.0 \text{ g/cm}^3$  and more in tapping 40 times is used as the graphite particles.

42. A method of preparing a carbon-based material for a negative electrode as claimed in claim 15, wherein natural graphite with the tapping density of  $0.9 \text{ g/cm}^3$  and more in tapping 20 times is used as the carbon-

based material, and the natural graphite is mechanically processed and pressing is performed thereon, and then a heat treatment is applied at a temperature within the range of 200 °C to 2300 °C, both inclusive.

43. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31 wherein natural graphite with the tapping density of 0.9 g / cm<sup>3</sup> and more in tapping 40 times is used as the carbon-based material, and the natural graphite is mechanically processed, and pressing is performed thereon, and then a heat treatment is applied at a temperature within the range of 200 °C to 2300 °C, both inclusive.

44. A method of preparing a carbon-based material for a negative electrode as claimed in claim 43, wherein the pressing is performed with a pressure of 1MPa and more.

45. A method of preparing a carbon-based material for a negative electrode as claimed in claim 31, wherein the method is used in a manufacturing process of a secondary battery where each of a negative electrode and a positive electrode has a structure in which a mixture containing an active material is formed on both sides of band-shaped collector and the battery is formed by stacking the negative electrode and the positive electrode with a separator made of a microporous film interposed therebetween, being spirally rolled a number of times.

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